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# Photonic bandgaps of conformally coated structures

## Abstract

Polymeric molds of the layer-by-layer photonic crystal can be economically synthesized with a microtransfer molding technique. The refractive indices of these molds are low, preventing formation of a photonic bandgap. We find that such molds can be conformally coated with higher-index material. Photonic band calculations find structures in which conformally coated layer-by-layer molds have complete bandgaps for both titania and silicon coatings. Large stop bands exist in the 001 stacking direction. Feasibility of experimental conformal coating of the molds has been demonstrated with a titania-coated polyurethane mold, which shows optical features in agreement with simulations of reflection and transmission.

## Keywords

Ames Laboratory, Physics and Astronomy, Electrical and Computer Engineering, Microelectronics Research Center

## Disciplines

Electronic Devices and Semiconductor Manufacturing | Materials Science and Engineering | Optics | Semiconductor and Optical Materials

## Comments

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# Photonic bandgaps of conformally coated structures

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Polymeric molds of the layer-by-layer photonic crystal can be economically synthesized with a microtransfer molding technique. The refractive indices of these molds are low, preventing formation of a photonic bandgap. We find that such molds can be conformally coated with higher-index material. Photonic band calculations find structures in which conformally coated layer-by-layer molds have complete bandgaps for both titania and silicon coatings. Large stop bands exist in the 001 stacking direction. Feasibility of experimental conformal coating of the molds has been demonstrated with a titania-coated polyurethane mold, which shows optical features in agreement with simulations of reflection and transmission. © 2005 Optical Society of America

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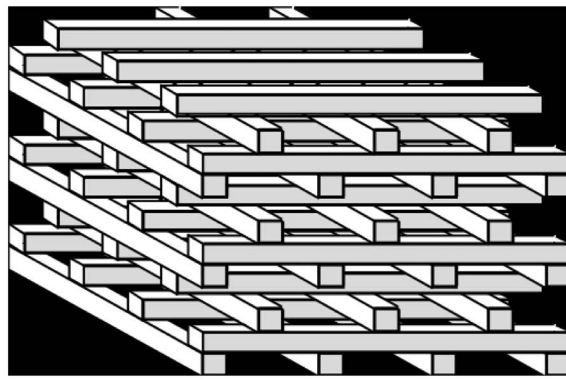
## 1. INTRODUCTION

Conformal coatings are widely used in the microelectronics industry, in which submillimeter-thickness coatings of protective materials are applied to electronic substrates or printed circuits. Such coatings provide mechanical and environmental protection, including corrosion resistance, electrical insulation, and protection against short circuits, to extend the life of electronic components and circuitry. Encapsulation also provides stress relief and electrical protection against contaminants. Coatings applied through spraying, dipping, and fluid flows are common. Polymeric coatings are extensively employed in medical devices or implants to ensure biocompatibility and biostability.<sup>1</sup> We demonstrate in this paper that conformal coating techniques can considerably improve the optical properties of photonic crystals.

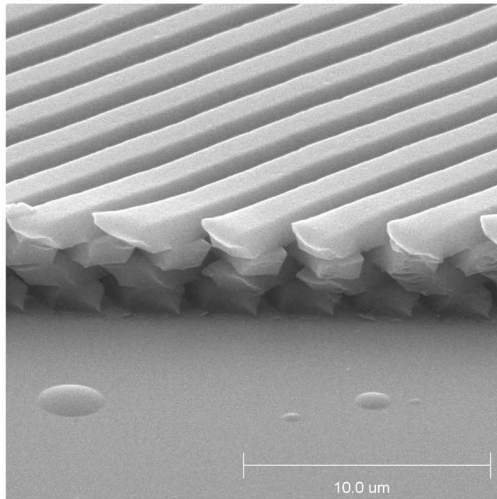
Three-dimensional photonic crystals with complete

bandgaps for omnidirectional propagation of electromagnetic waves have immense potential for devices in fiber-optics-based telecommunications applications, single-mode waveguides, channel add-drop filters, catalysis, and control of spontaneous emission. The three-dimensional layer-by-layer structure designed at Iowa State<sup>2-4</sup> with a large robust bandgap has been fabricated with bandgap wavelengths of 10,<sup>5</sup> 1.5,<sup>6</sup> and 1.3  $\mu\text{m}$ <sup>7</sup> by state-of-the-art semiconductor fabrication methods. Such photonic crystals with precise submicrometer features have excellent optical performance over small areas. It is desirable to investigate alternative economical fabrication methods in modest laboratory facilities that can rapidly synthesize large-area photonic crystals operating at optical and near-infrared wavelengths.

An alternative fabrication method is the microtransfer molding method<sup>8</sup> in which an elastomeric mold is created



(a)



(b)

Fig. 1. (a) Schematic figure of the original layer-by-layer photonic crystal before coating, with rectangular dielectric rods in each layer. (b) Scanning electron microscope image of the experimentally fabricated four-layer mold structure of polyurethane bars, viewed with a cross-sectional cut at  $45^\circ$  to the axis of the rods. This mold structure is the starting point for the conformal coating.

out of polymer material and a master. The elastomeric mold can be filled with a polymer such as polyurethane, and the single-layer polyurethane pattern can be transferred to a substrate.<sup>9</sup> By repeating this process for successive layers, one can easily fabricate a large-area three-dimensional photonic crystal consisting of low-index polymeric material. Owing to the low index of this polymeric material ( $n \sim 1.5$  as in polyurethane), such photonic crystals cannot exhibit a three-dimensional bandgap. One route to improve the index contrast is to infiltrate a higher-index ceramic material (such as titania with  $n \sim 2.6$ – $2.7$ ) into the structure and then remove the polymeric mold by calcination at high temperature. We propose an alternative method to improve the contrast of the polymeric template by coating the polymeric crystal with a conformal coating of a higher-index material such as titania. Conformal coatings of higher-refractive-index semiconductors such as silicon or III–V materials can be achieved. Such coated structures with three dielectric materials have not been investigated experimentally or theo-

retically. In this paper we calculate the photonic bandgaps of coated rods and experimentally synthesize a coated photonic crystal.

## 2. COATED STRUCTURES

The starting point for the calculations is the layer-by-layer (woodpile) structure<sup>2–4</sup> with lower-refractive-index rods (width  $w$ , height  $h$ , spacing  $d$ ) and a periodicity of four rods in the  $z$  direction shown in Fig. 1(a). Each layer consists of a one-dimensional array of rectangular rods. Successive layers are rotated by  $90^\circ$  relative to each other. Second-neighbor layers are parallel to each other but shifted by  $d/2$ .

This structure has been fabricated using microtransfer mold techniques. A flexible elastomeric mold is created out of polymer material and a master. The elastomeric mold is filled with a polymer such as polyurethane, and the single-layer polyurethane pattern was transferred to a substrate<sup>9</sup> such as glass or silicon. The mold was peeled off, the substrate retaining the pattern. The mold was re-filled with polymer, and we formed the second layer by rotating the substrate by  $90^\circ$  relative to the elastomeric mold and transferring the polymer bar pattern on top of the first layer. This process was repeated for the third layer, which was aligned so that the third-layer bars were in between the first-layer bars. Moiré fringe alignment was utilized for achieving large well-aligned areas.<sup>10</sup> By our repeating this process, a large-area three-dimensional photonic crystal consisting of four layers of low-index polymeric material was easily fabricated [Fig. 1(b)]. When this structure is conformally coated [schematically shown

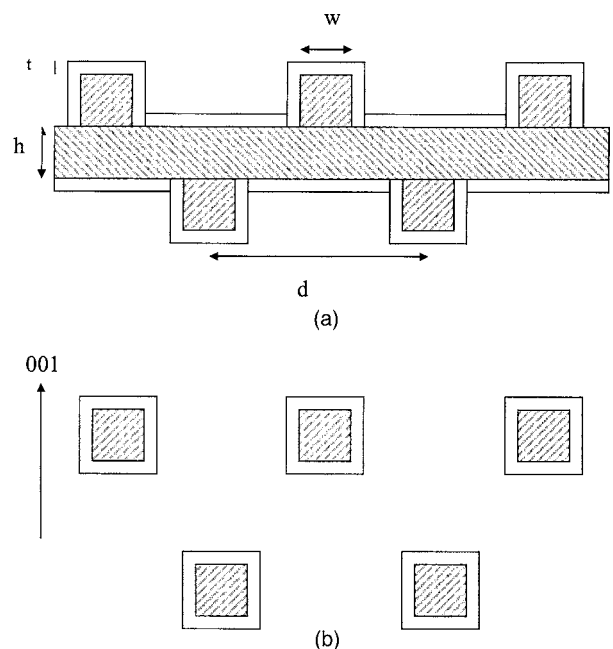


Fig. 2. (a) Schematic figure of a conformally coated structure showing lower-refractive-index molds with bar width  $w$ , coated with higher-refractive-index material to a thickness  $t$ . The bar separation  $d$  and mold height  $h$  are unaltered by the coating. A cross section ( $xz$  plane) showing adjacent bars in the  $z$  direction are shown. (b) A conformally coated structure with another cross section that does not include second- and fourth-layer bars is shown.

in Figs. 2(a) and 2(b)], the exposed surfaces of the lower-refractive-index bars are uniformly coated with higher-refractive-index material (with a thickness  $t$ ), resulting in a multiply connected geometry. The spacing ( $h$ ) between bars in the  $z$  direction is unaltered. A cross section of the coated structure in a plane containing neighboring bars in the  $z$  direction shows the coating filling in from the vertical faces of bars in layer 1 and from the horizontal faces of bars in the next layer, layer 2. When the structure is sliced in a plane that does not contain the bars from layer 2, we find the coating uniformly encapsulates the faces of the bars. As the coating thickness  $t$  increases, the filling fraction of the higher-index coating increases from both of these contributions. Although photonic band calculations have been performed extensively for the layer-by-layer structure, no band calculations are available for such conformally coated structures with low-index cores. We investigate optimal geometries of conformal-coated structures with photonic band calculations and describe experimental fabrication of such conformally coated lattices.

### 3. RESULTS

The photonic band structure was calculated with the standard vector-wave plane-wave expansion method<sup>2,3</sup> for describing the photonic bands in a periodic dielectric structure. The electric and magnetic fields are superposition of plane waves. Typically, calculations involve diagonalization of matrices of sizes 850–900, which lead to good convergence. The coated rod structures were discretized in real space from which fast Fourier transforms generated Fourier components of the dielectric matrix.

We start with a structure with an inner polymeric core of refractive index  $n=1.5$  and bar width  $w$  and increase the thickness  $t$  of the titania coating ( $n=2.7$ ). The full bandgap for coated structures was calculated (Fig. 3) for different starting polymeric templates of width  $w$ . The initial uncoated polymeric template ( $t=0$  in Fig 2) does not have complete bandgaps but does have small stop bands (of width  $<6\%$ ) in the 001 stacking direction. As the high-index coating thickness  $t$  is increased, the 001 stop band increases, and the structure evolves toward a full bandgap (Fig. 3). The initial thin molds with a filling fraction or width/separation ratio  $w/d$  of 4.5%–9% do not have a full gap, but, as they are coated with titania ( $n=2.7$ ), a full bandgap for all directions of propagation emerges and reaches a maximum value of 2%–3%. Coating thicknesses  $t/d \sim 0.1$  are optimal for these structures. The thicker molds show some improvement when coated but no full bandgap. The dielectric filling ratio is  $\sim 29\%$  at the best performing geometry. Multiply connected structures with overlapping rods in the layer-by-layer structure have previously shown improved bandgaps over the simple stacked layer-by-layer structure.<sup>11</sup> In the limit of  $w=0$ , there is no mold, and the structure is composed of a high-index coating; the complete bandgap ratio reaches the limiting value of 10% for refractive index coatings of  $n=2.7$ .

The gap in the 001 stacking ( $z$ ) direction is large (Fig. 3) for all the coated structures and reaches a maximum near 15%. Transmission in the  $z$  direction is expected to

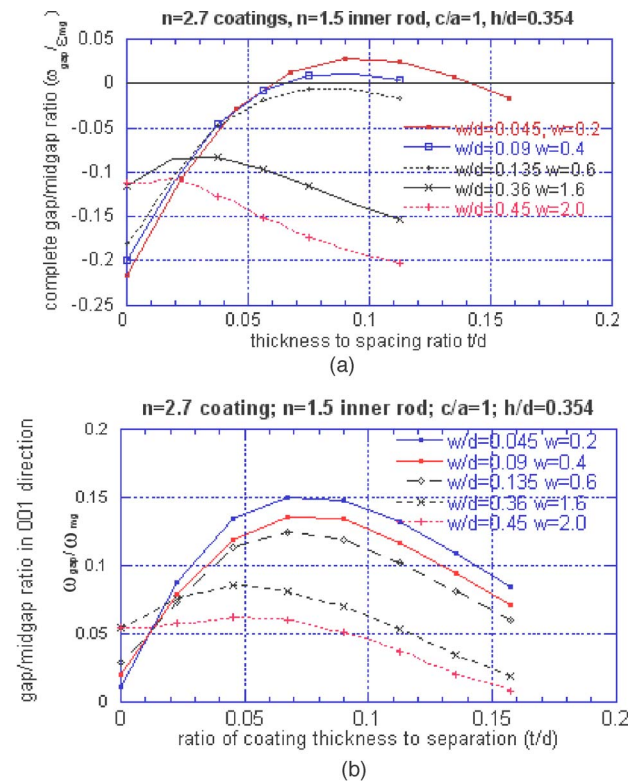


Fig. 3. (Color online) (a) Dimensionless ratio of complete photonic bandgaps for all directions of propagation ( $\omega_{\text{gap}}$ ) to the mid-gap frequency ( $\omega_{\text{midgap}}$ ) for conformally coated molds with an inner polymeric core of refractive index 1.5 and a higher-index titania coating ( $n=2.7$ ) as a function of the coating thickness  $t$ . Each curve corresponds to a fixed width  $w$  of the polymeric bar. Positive gap/midgap ratios correspond to complete bandgaps. (b) Stop bands in the 001 direction for polymeric bar molds coated with titania following the same conventions as in (a).

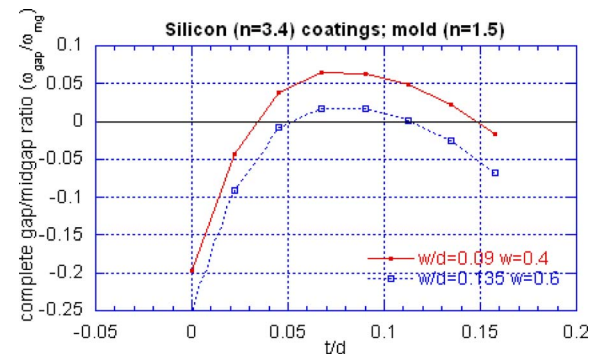


Fig. 4. (Color online) Ratio of complete bandgap to the midgap frequency for a silicon-coated layer-by-layer mold as a function of thickness  $t$  of the silicon coating for two widths  $w$  of the mold bars. The refractive indices were 3.4 for silicon and 1.5 for the polymeric mold.  $w/d$  is the ratio of the mold bar width to the bar separation. The conventions follow those of Fig. 2.

show a sizable stop band for the overcoated structures with thick coatings, even when no full gap is found. Such robust stop bands may already be useful for optical applications.

Higher-performing conformally coated structures can be achieved by using higher-index coatings such as silicon by using deposition techniques such as sputtering. For thin molds ( $w/d=0.09$ ), a complete bandgap with a mag-



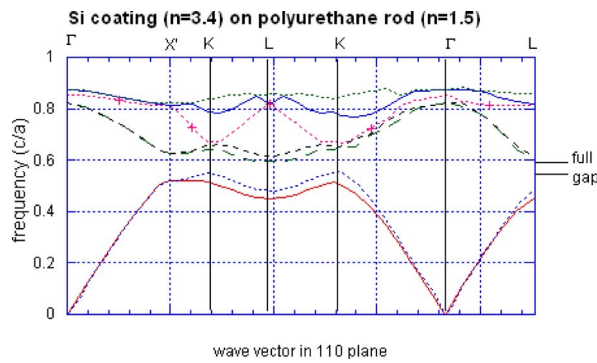


Fig. 5. (Color online) Photonic band structure of the conformally coated structure with silicon coatings ( $n=3.4$ ) on a polyurethane rod template ( $n=1.5$ ). The coating thickness  $t/d=0.068$ , corresponding to the best performing structure.

nitude of 5%–6% can be achieved (Fig. 4) with moderate coating thicknesses (thickness ratios  $t/d \sim 0.07$ – $0.09$ ). For slightly thicker molds ( $w/d=0.135$ ), the complete gap is reduced to  $\sim 2\%$  (Fig. 4). Hence such high-index conformal coatings of economically fabricated molds can be viable for low-cost large-area photonic crystals with complete bandgaps. The band structure (Fig. 5) demonstrates a complete bandgap over the Brillouin zone with the valence band (band 2) maximum at K and the conduction band (band 3) minimum at L, similar to the layer-by-layer photonic band structure.

A further improvement can be realized by removal of the low-index polymeric mold by calcinations, resulting in a structure with higher dielectric contrast and a gap/midgap ratio of 8.4% for silicon coatings, with similar thin cores  $w/d=0.045$ . The magnitude of the gaps is limited by the coating thickness in the 001 direction, which causes overlaps of the rods in the  $z$  direction. We also anticipate that higher gaps can be achieved for nonuniform coating.

#### 4. EXPERIMENTAL SYNTHESIS AND OPTICAL MEASUREMENTS

We experimentally demonstrate the feasibility of conformal coatings by fabricating a four-layer conformally coated structure (Fig. 6). We first synthesize a four-layer polyurethane mold template with a bar separation of  $2.5 \mu\text{m}$ , which was easily achieved over a large area by the microtransfer molding method.<sup>9,10</sup> The layer-by-layer stacking of the template was achieved by the moiré-fringe-based alignment method. For these preliminary studies, we experimentally employed a moderate polyurethane bar width  $w \sim 1.4 \mu\text{m}$  and conformally coated the structure with titania to a coating thickness of  $0.45 \mu\text{m}$ , using the atomic-layer deposition method. Since the processing temperatures are  $\sim 100^\circ\text{C}$ , the titania remains in the anatase phase rather than the higher-index rutile phase. The resulting coated structure (Fig. 6) demonstrates a uniform coating of each polymeric rod, including on the back side of each rod, together with rounded corners. Since a high-filling-ratio mold ( $w/d \sim 0.56$ ) was employed, the structure is overfilled. The synthesis demonstrates that conformal coatings may be achieved experimentally, and a future direction is to evolve toward thinner polymeric molds and higher-index coatings.

The optical properties of the conformally coated structure were measured with a Fourier transform infrared spectrometer and compared with the uncoated polyurethane mold (Fig. 7). There is transmission between 2 and  $4.5 \mu\text{m}$  for both the mold and the conformally coated structure accompanied by significant reflection ( $R$ ) for the coated structure. For wavelengths above  $5 \mu\text{m}$ , the transmission ( $T$ ) for both structures is low, since the substrate (Corning No. 1 cover glass) absorbs strongly in this long-wavelength range. The glass absorption causes the steep transmission edge at  $5 \mu\text{m}$ . Moreover, some minor changes on the reflectance due to the absorption bands of polyurethane are observed at around  $5.8$  and  $6.8 \mu\text{m}$ . At short wavelengths below  $4 \mu\text{m}$ , substantial diffraction occurs, causing the specular transmission to be low (Fig. 7). After accounting for diffraction, we find the absorption is low for wavelengths below  $4 \mu\text{m}$ . The reflection peaks at  $9$  and  $11 \mu\text{m}$  are present for the glass substrate and are the well-known absorption peaks of silica from the glass where the  $\text{Im}(\epsilon)$  has maxima.

However, the reflective peak between  $6$  and  $7 \mu\text{m}$  for the conformally coated structure is not present for the four-layer mold. The position of this feature is consistent with the expected stop band in the 001 direction for such an overcoated structure, taking onto account the decrease of the refractive index of anatase titania to  $\sim 1.9$  in this wavelength range.<sup>12</sup>

This interpretation is supported by rigorous  $S$ -matrix (scattering matrix) simulations<sup>13</sup> performed on the multilayer structure. Each layer of our structure can be decomposed into sublayers in which the structure simplifies to a one-dimensional grating. Within each such grating layer, Maxwell's equations can be rigorously solved in a plane-wave basis set. The simulations utilize the experimental values of  $n_1 + in_2$  for titania and polyurethane. Experimental values of  $n_1 + in_2$  for the glass substrate were inferred through measurement. Simulated reflection and

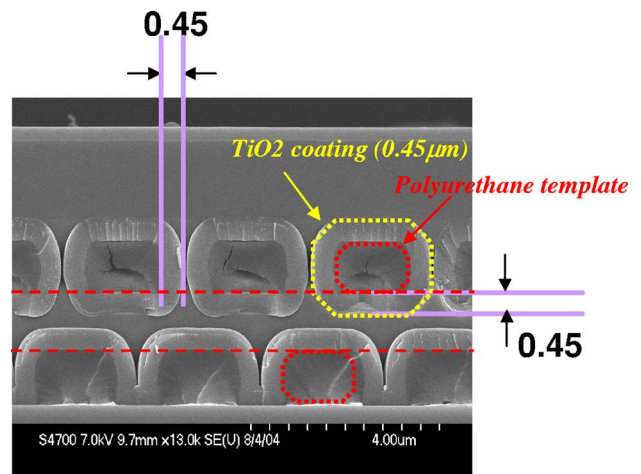


Fig. 6. (Color online) Experimentally synthesized conformal titania coating of a four-layer polyurethane bar template using the atomic-layer deposition method. A titania coating of  $0.45 \mu\text{m}$  was achieved for a template with bar separation of  $2.5 \mu\text{m}$  and bar width of  $1.4 \mu\text{m}$ . In the scanning electron micrograph figure the structure is sliced in a plane at  $90^\circ$  to the first and third layers. The coating (lighter region) of titania is  $0.45 \mu\text{m}$  thick for an inner bar (dark region) width of  $1.4 \mu\text{m}$ . The dotted lines show the outline of the recessed second layer before the conformal coating.

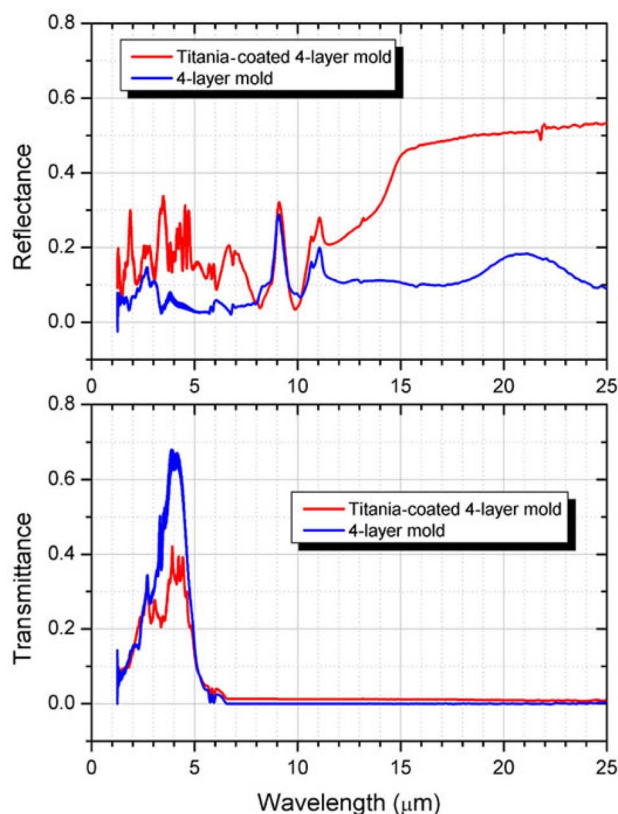


Fig. 7. (Color online) Reflection and transmission measurements for the four-layer conformally coated structure on a  $150\text{ }\mu\text{m}$  thick glass substrate, compared with measurements for the uncoated polyurethane mold on the same glass substrate.

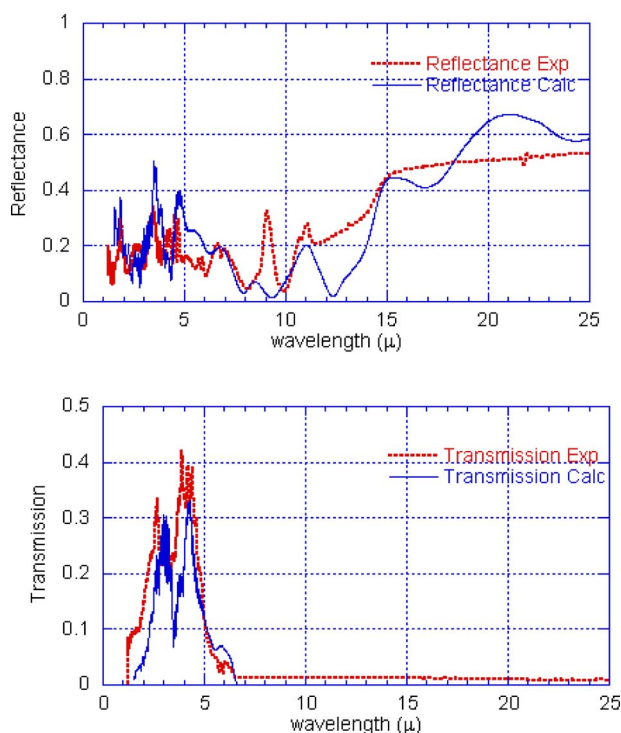


Fig. 8. (Color online) Simulated reflectance and transmission from *S*-matrix calculations for the four-layer conformally coated mold, compared with the experimental data of Fig. 6. The calculations used a bar width  $w=1.3\text{ }\mu\text{m}$ , coating thickness  $t=0.45\text{ }\mu\text{m}$ , and bar separation  $d=2.5\text{ }\mu\text{m}$ .

transmission for the coated structure on a  $150\text{ }\mu\text{m}$  thick glass substrate show (Fig. 8) transmission peaks near 3 and  $4\text{ }\mu\text{m}$  in good agreement with experiment, followed by low transmission above  $6\text{ }\mu\text{m}$ , due to the absorption in glass. The simulated reflectance shows a weak peak between 6 and  $7\text{ }\mu\text{m}$  in the same position as in the measurement. Simulated reflected peaks near 3, 5, and  $11\text{ }\mu\text{m}$  are in good agreement with data, although there is some difference in the  $9\text{--}10\text{ }\mu\text{m}$  range between calculation and experiment. We interpret the reflective peak between 6 and  $7\text{ }\mu\text{m}$  that is accompanied by negligible transmission as occurring from a weak photonic stop band in the stacking direction. This feature can be enhanced for higher-refractive-index contrasts or thinner coatings. Conformal coatings may improve the dielectric contrast in other low-refractive-index photonic crystals generated in photorecist by holographic methods<sup>14</sup> or direct laser writing.<sup>15</sup>

## 5. CONCLUSIONS

Conformal coatings can improve the optical contrast in low-refractive-index photonic crystals such as submicrometer-scale structures generated in polymeric materials by microtransfer molds. Conformal coatings may be helpful in improving low-index photoresist lattices that were produced holographically. We find geometries of the conformal-coated structures that lead to complete photonic bandgaps for both titania and silicon coatings of lower-index polymeric molds. A sizable stop band in the stacking direction is present even when a complete bandgap cannot be achieved. Conformal titania coatings of a four-layer structure were achieved with the atomic-layer deposition method. Optical properties reveal features consistent with a photonic stop band. Improved photonic bandgaps will be achieved in future research by using thinner polymeric molds and higher-refractive-index coatings.

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